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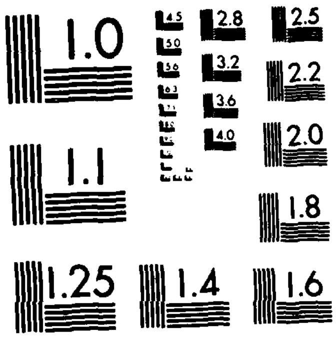

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One of the long range goals of this project has been to produce a photoexcited exciton system which displays quantum statistics. We are attempting to find a method to produce excitions in a superconducting or superfluid state.

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Technical Report: AFOSR Grant 79-0124.

MEASUREMENT OF HIGH MOBILIITES AND STRAIN CONFINEMENT OF LONG-LIVED FREE EXCITONS IN Cu_2O.

By J. P. Wolfe, Professor of Physics
Principal Investigator

One of the long range goals of this project has been to produce a photoexcited exciton system which displays quantum statistics. ~~We are attempting to find a~~ A method is to produce excitons in a superconducting or superfluid state. One of the prime difficulties in achieving this goal is that excitons in direct gap semiconductors generally have very short lifetimes, of order 1 nanosecond, which is not long enough to permit their kinetic energies to come into equilibrium with the lattice temperature. On the other hand, the direct gap semiconductor exhibits excitons with very small Bohr radii advantageous to observing quantum statistics. To circumvent the lifetime problem stated above, ~~we have concentrated on~~ high purity crystals of Cu_2O, which recently were shown to display excitonic lifetimes in the microsecond range.

We have discovered that it is possible to transport excitons millimeter distances in a high purity crystal of Cu_2O at low temperatures, $T \approx 2$ K. The experiments imply that the paraexciton in this crystal has an extremely high mobility — of order 10^6 cm^2/Volt-sec. This is the first measurement of an excitonic drift mobility in a direct gap semiconductor. Also, we have found that it is possible to confine these excitons in a strain potential well within the bulk of the crystal. To do this, it was necessary to conduct a

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detailed study of the energy of the paraexciton as a function of crystal stress. Our data and a subsequent theory which agrees with these results will be published in Solid State Communication. (A preprint of the accepted publication is attached.) The principle result in our strain confinement experiments is that the excitons, produced resonantly by a tunable dye laser, exhibit thermal equilibrium over the entire volume which they occupy. Our preliminary findings on strain confinement and high mobility of excitons in Cu_2O will be presented at the March Meeting of the American Physical Society, 1983, and the corresponding abstracts are attached. We are presently attempting to extend these measurements to higher exciton densities and lower temperatures in order to observe the onset of quantum statistics. We hope to demonstrate Bose condensation of this excitonic system, which may be accompanied by superfluid transport.

STRESS DEPENDENCE OF THE PARAEXCITON IN Cu_2O

by

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ABSTRACT

The energy shift of the paraexciton in Cu_2O is measured by detecting its luminescence. The stress dependence, which differs significantly from that of the orthoexciton, agrees well with a theoretical calculation.

Cu_2O offers a rather unique system for the study of Wannier-Mott excitons. It exhibits a large variety of optical transitions of excitonic origin (first class, second class, phonon-assisted, quadrupole, etc.) which are in excellent agreement with the theory of Elliot.[1] The lowest excitonic state corresponds to the n = 1 term of the yellow series which are formed from a Γ_6^+ conduction band electron and a hole from the upper Γ_7^+ valence band. Electron-hole exchange interaction splits the n = 1 level into a triply-degenerate Γ_5^+ orthoexciton (X_0 = 2.034 eV at 2 K) and a lower lying, singly-degenerate paraexciton Γ_2^+ (X_p = 2.022 eV at 2 K). An attractive feature or the paraexciton resides in the fact that it remains the lowest electronic excitation of the system even under conditions of strong particle interaction. Contrary to most other highly-excited semiconductors, biexcitons and electron-hole liquid are not energetically favorable.

Recently, Waters et al.[2] and Trebin et al.[3] have reported an extensive study of the stress dependence of the orthoexcitons in Cu_2O. Reasonably good agreement was found between experimental results and the model of Trebin et al. The orthoexciton degeneracy is partially removed under the action of applied pressure in the [100] direction, with the lowest component shifting down in energy by an amount which becomes comparable, at 3.0 kbar, to the unperturbed crystal ortho-para splitting. The question arises then of what the energy position of the paraexciton does under similar conditions. A previous study by Kreingold and Makarov[4] indicated that, unlike the orthoexciton, the paraexciton energy is unchanged for stresses below 1.5 kbar. In this paper, we report an experimental study of the paraexciton over a continuous range of stress values and find a significant effect. We compare the results to theory, using the model of reference 3.

The measurements were performed in high-quality single crystals of

natural growth which were cut and polished in the form of cubes with lateral dimensions of 1.7 mm. Pressure was applied along the [100] axis using a spherical contact surface from either a stainless steel or glass stresser of radius 4 mm. As is well known[5], the resulting Hertzian strain distribution leads to a maximum shear strain inside the sample. In our case the maximum strain occurred approximately 0.3 mm below the contact surface at a maximum applied pressure of 3.0 kbar. The Hertzian stress geometry has the advantage of allowing high stresses without breaking the crystal, a limitation encountered in previous experiments.[4]

In the experiments described here, only that part of the sample corresponding to the minimum energy of the stress-induced potential well was excited using a CW dye laser tuned to the locally-downshifted threshold of the phonon-assisted orthoexciton absorption continuum. The exciton luminescence emitted from the well was viewed along a [110] axis and recorded as a function of applied pressure using a 1/2 meter spectrometer in conjunction with an OMA detector (spectral resolution approximately 0.5 Å). For every applied pressure, the location of the excitation was adjusted to the bottom of the strain well, which optimized the signal emitted from the corresponding volume. Values for the zero-stress limit were obtained by surface excitation and detection.

Luminescence spectra recorded at T = 2.0 K as a function of applied pressure are presented in Figure 1. The pressure calibration was obtained from the orthoexciton energy position, using the theory of Trebin. This calibration was consistent with an estimate of the shear strain maximum for the Hertzian geometry with our experimental parameters. Figure 2 is a higher-gain recording of the zero-stress and 2.8 kbar spectra, giving an identification of the various phonon replicas. Besides the direct quadrupole

recombination of the lowest twofold-degenerate orthoexciton (of symmetry Γ_5^+), X_o, and its Γ_{12}^- (108 cm^{-1}) phonon replica, one observes the appearance of the direct paraexciton X_p (2.015 eV at 2.8 kbar) and its weak phonon replica (X_p - Γ_{25}^-). A rapid increase in intensity of the direct paraexciton luminescence with stress is clearly apparent in the data of Figure 1, as was reported previously in the lower-stress experiments.[4] This intensity "ignition" was attributed to a mixing with the exciton states from the green series. In Figure 3, we plot the position of the paraexciton versus pressure, together with results of the calculated dependences for both ortho and paraexcitons. As can be seen, a very good agreement is found between experiment and theory. The paraexciton shift is quite small below 1.5 kbar but increases significantly above this stress.

Finally, we briefly comment on some possible implications of the results. The paraexciton remains the lowest energy state of the crystal, even in the presence of the stress. This means that it should be possible to use the Hertzian stress distribution geometry to attract and confine long-lived paraexcitons in the well. Experiments along these lines are in progress. Also, it may be noted that the ortho-para splitting diminishes with applied pressure. This splitting is a crucial parameter in recent models which attempt to explain the striking temperature dependence of ortho and paraexciton intensities.[6,7] In particular, above 0.8 kbar, the splitting becomes less than the lowest energy optical phonon of the crystal (Γ_{25}^- = 85 cm^{-1}), which has been postulated to mediate the ortho-para conversion. Thus the properties of ortho and paraexcitons under stress may give some further insights into the basic mechanisms of ortho-para conversion.

This work was supported in part by the Air Force Office of Scientific Research under the Grant AFOSR79-0124 and by a scholarship of the Deutsche

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Figure Captions

Figure 1. Luminescence spectra as a function of applied stress recorded at T = 2.0 K. X_o is direct recombination of lowest strain-split orthoexcitons. X_p is direct recombination of paraexcitons. Stress calibration was obtained from orthoexciton line using the theory of Trebin et al. (Reference 3).

Figure 2. Higher gain plots of zero-stress and highest stress spectra of figure 1 showing various phonon replicas.

Figure 3. Ortho and paraexciton energy as a function of stress. Solid lines are Trebin's theoretical curves for ortho and paraexcitons. Circles are experimental data points for paraexciton using position of orthoexciton to calibrate the stress implied.

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*Permanent Address: Groupe de Physique des Solides de l'Ecole Normale Supérieure, 75005 Paris, France.

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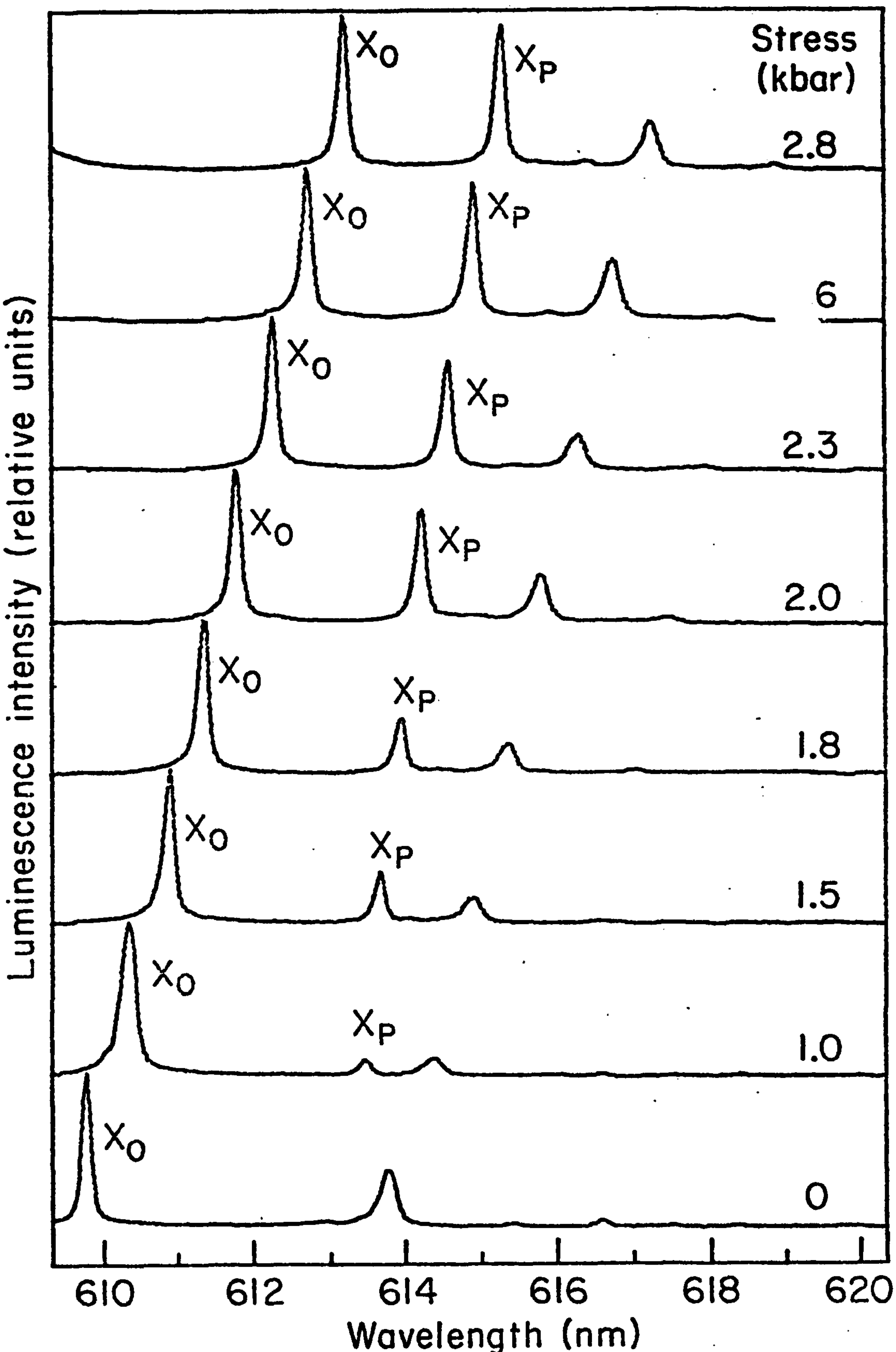
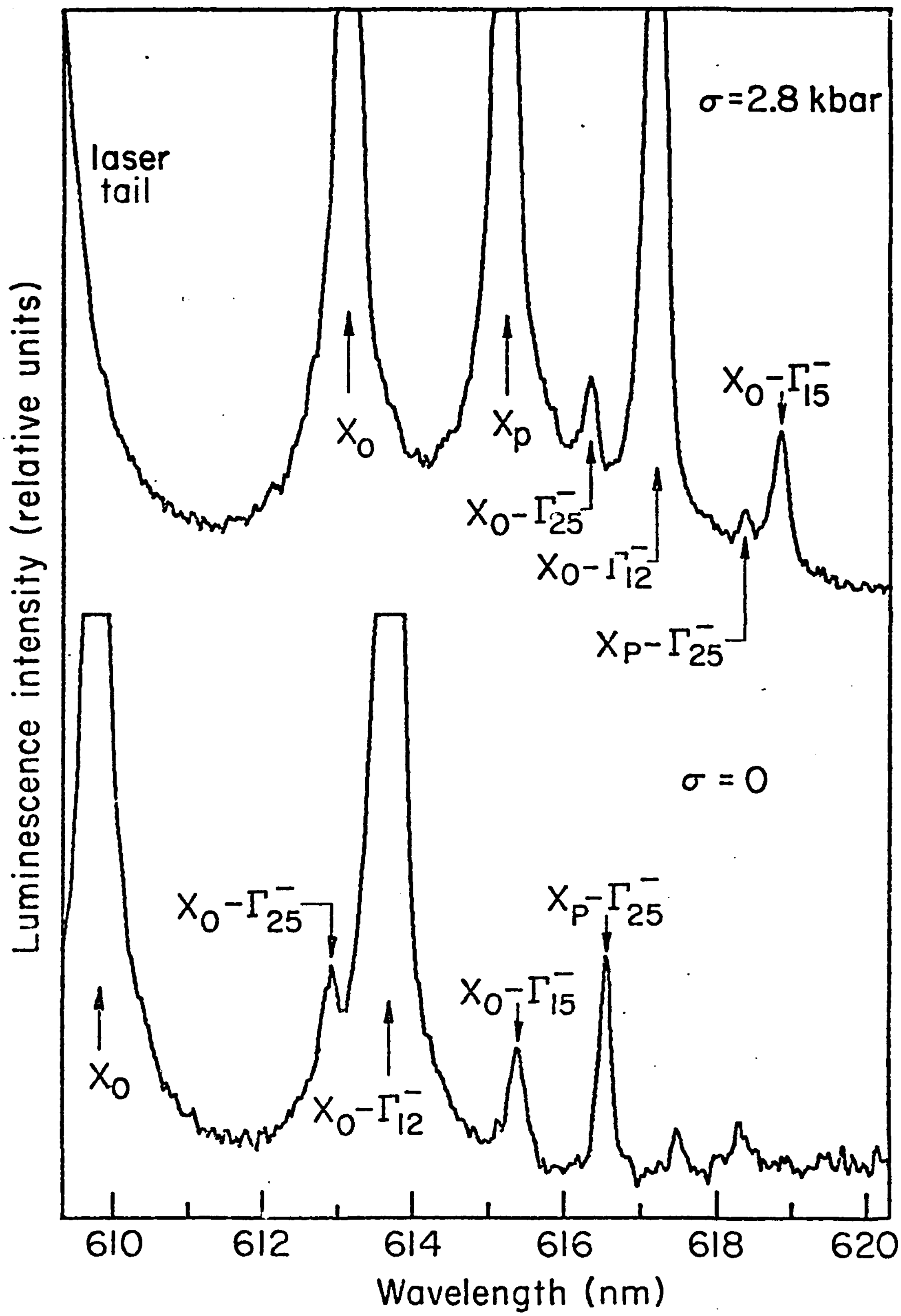

Stress (kbar)
2.8
6
2.3
2.0
1.8
1.5
1.0
0
X_O
X_P
Luminescence intensity (relative units)
610
612
614
616
618
620
Wavelength (nm)

Figure 1



σ=2.8 kbar
laser tail
X_0
X_p
$X_0-\Gamma_{15}^-$
$X_0-\Gamma_{25}^-$
$X_0-\Gamma_{12}^-$
$X_P-\Gamma_{25}^-$
σ = 0
$X_0-\Gamma_{25}^-$
$X_P-\Gamma_{25}^-$
$X_0-\Gamma_{15}^-$
X_0
$X_0-\Gamma_{12}^-$
Luminescence intensity (relative units)
610
612
614
616
618
620
Wavelength (nm)

Figure 2

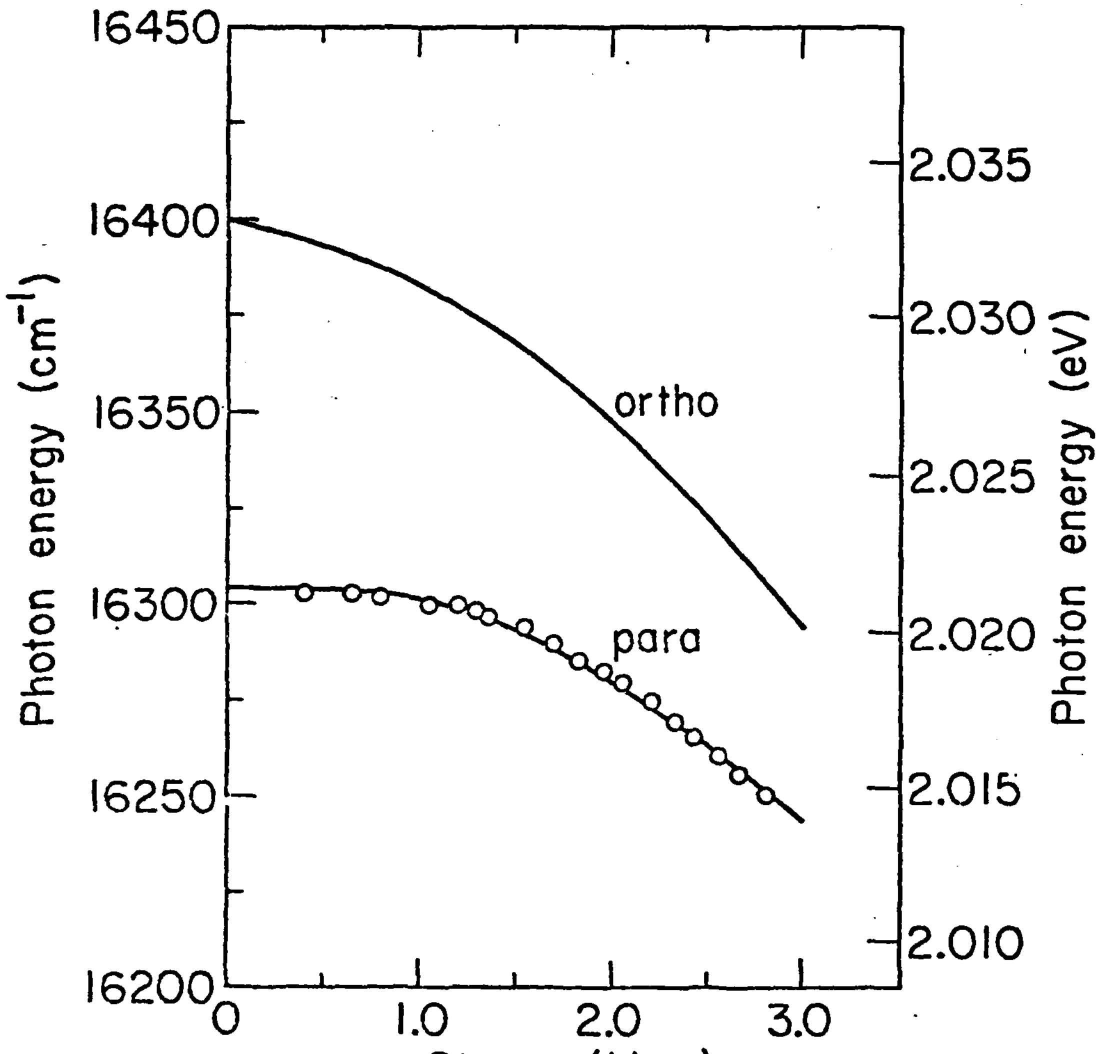

16450
16400
16350
16300
16250
16200
2.035
2.030
2.025
2.020
2.015
2.010
Photon energy (cm^{-1})
Photon energy (eV)
ortho
para
0
1.0
2.0
3.0
Stress (kbar)

Figure 3

Abstract Submitted
for the March 19⁸³ Meeting of the
American Physical Society

Physics and Astronomy
Classification Scheme
Numbers 71.35.+z
78.55.-m

Suggested title of session
for this paper
Excitons and Electron
Droplet

Strain-Confinement of Free Excitons in Cu_2O.†

J. P. Wolfe, D. P. Trauernicht, and A. Mysyrowicz*,
Physics Dept. and Materials Research Laboratory, University of Illinois, Urbana, IL 61801.
— We have succeeded in strain-confining free excitons in a direct-gap semiconductor at liquid He temperatures. The excitons are produced in high-purity natural-growth Cu_2O by CW excitation with Ar^+ or tunable dye-laser light. A parabolic potential well is produced by a Hertzian stress configuration. Luminescence from both para- and ortho-excitons is detected. As a result of their long lifetime, the paraexcitons exhibit equilibrium thermodynamics at the lattice temperature, as determined by their spatial and spectral distributions. In particular, the volume occupied by these excitons shows a $T^{3/2}$ dependence, characteristic of the classical regime.

† Supported by Air Force Office of Scientific Research Grant 79-0124.
* Permanent address: GPS-ENS, Paris, France, laboratoire associé au CNRS.

(x) Prefer Standard Session

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Abstract Submitted
for the March 1983 Meeting of the
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Physics and Astronomy
Classification Scheme
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78.55.-m
72.20.Fr
72.10.Di

Suggested title of session
for this paper
Excitons and Electron-
Hole Droplets

High Mobility of Paraexcitons in Cu_2O.†

D. P. Trauernicht, A. Mysyrowicz* and J. P. Wolfe,
Physics Dept. and Materials Research Laboratory, University of Illinois, Urbana, IL 61801. — Using the strain-gradient method, we have observed a macroscopic drift of free excitons in high-purity natural-growth Cu_2O. Paraexcitons produced by Ar^+ laser excitation at the crystal surface are transported over macroscopic distances (>5mm) into the crystal by a calibrated energy gradient. Both the motive force and drift velocity are measured, giving a direct determination of the excitonic drift mobility. Extremely high mobilities — of order 10^6 cm^2/V-sec — are found at liquid helium temperatures. The temperature dependence of the exciton mobility suggests that the momentum damping time is dominated by carrier-phonon scattering.

† Supported by Air Force Office of Scientific Research Grant 79-0124.

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